

(12) UK Patent Application (19) GB (11) 2 374 948 (13) A

(43) Date of A Publication 30.10.2002

(21) Application No 0110299.5

(22) Date of Filing 27.04.2001

(71) Applicant(s)

Ashe Morris Limited
(Incorporated in the United Kingdom)
6 Christchurch Crescent, RADLETT, Hertfordshire,
WD7 8AH, United Kingdom

(72) Inventor(s)

Robert Ashe
David Morris

(74) Agent and/or Address for Service

Bawden & Associates
66A High Street, HARPENDEN, Hertfordshire,
AL5 2SP, United Kingdom

(51) INT CL⁷

B01J 19/00 , G05D 23/00

(52) UK CL (Edition T)

G3R RBQ59
B1X X16

(56) Documents Cited

US 5762879 A
Collect. Czech. Chem. Commun. Vol 47 No 2, 1982, pp
446-453 Temperature control in chemical reactor

(58) Field of Search

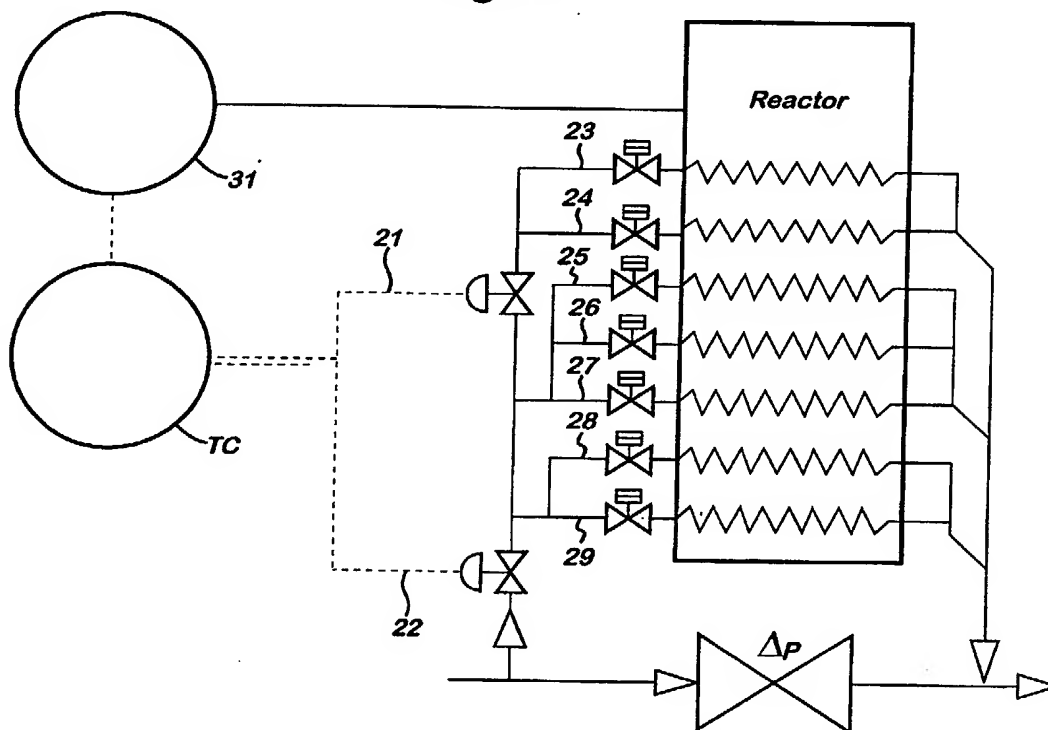
UK CL (Edition S) B1X X16 X20 , G3R RBQ59
INT CL⁷ B01J 19/00 , B01L 7/00 , F28F 13/00 27/00
27/02
ONLINE: EPODOC, JAPIO, WPI, DIALOG DIALINDEX
CHEMENG

(54) Abstract Title

Reaction system with variable heat transfer surface area

(57) The reaction system comprises a process (reaction) fluid and a heat transfer fluid which passes in a conduit through the process fluid. The heat transfer surface area of conduit available to the process fluid may be varied according to the heat generated or absorbed by the reaction, as determined by the temperature change in the heat transfer fluid across the reaction and the mass flow of the heat transfer fluid through the conduit. The conduit may comprise a number of pipes or coils which can be selected by operation of valves 23-29.

Fig. 6



At least one drawing originally filed was informal and the print reproduced here is taken from a later filed formal copy.

This print takes account of replacement documents submitted after the date of filing to enable the application to comply with the formal requirements of the Patents Rules 1995

GB 2 374 948 A

Fig. 1

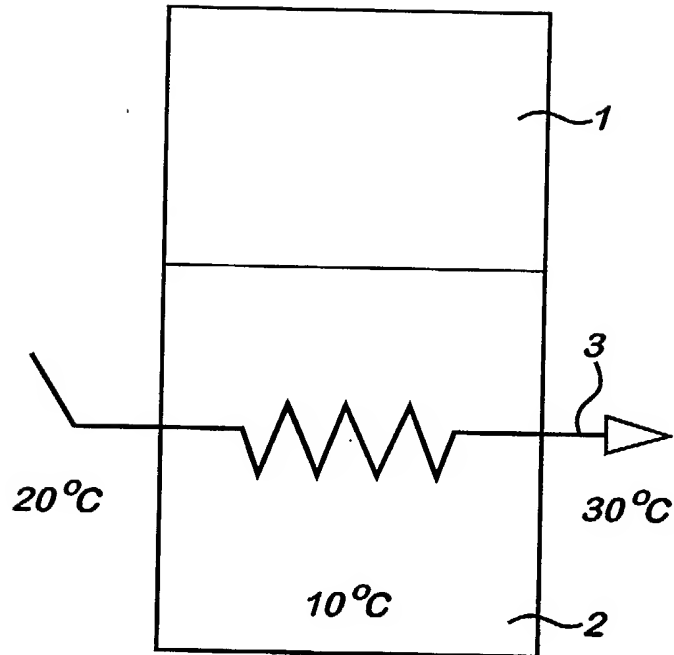
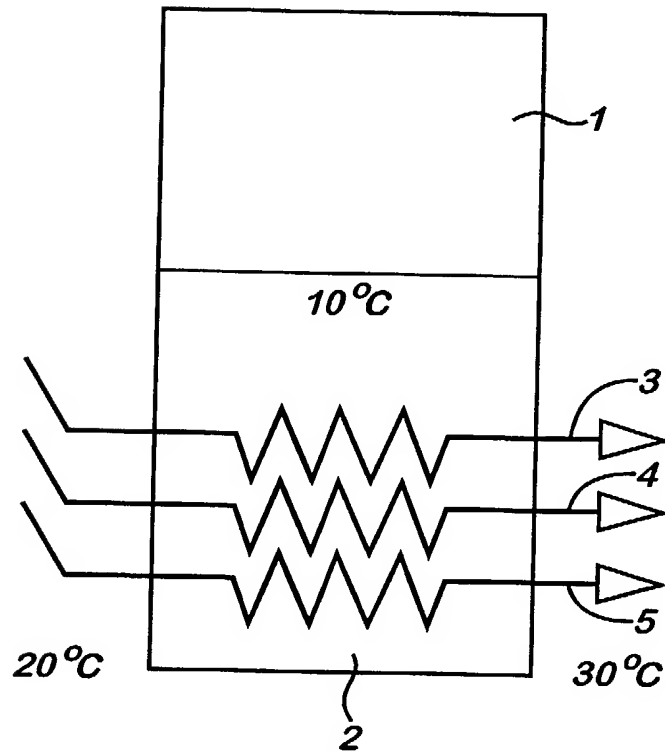


Fig. 2



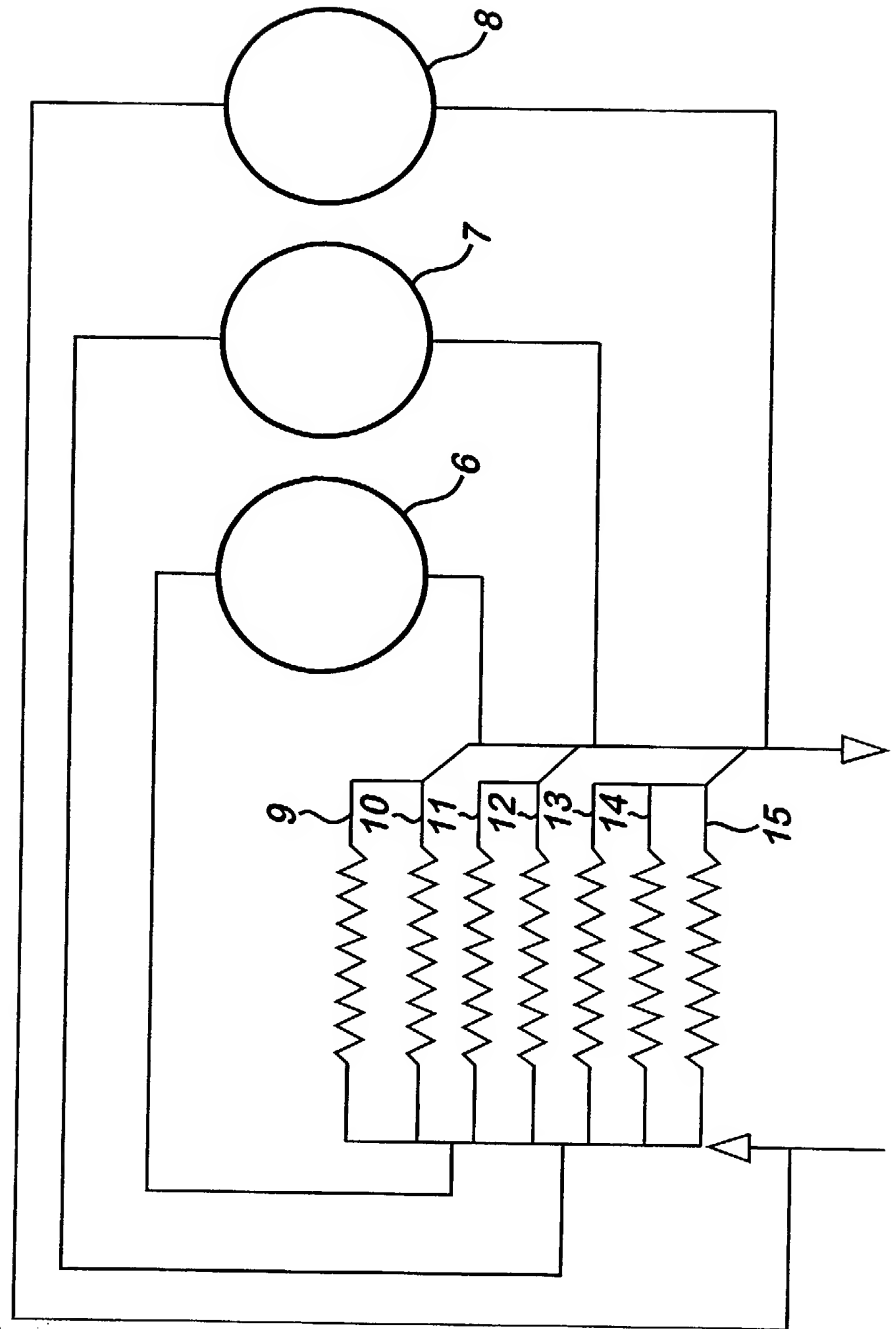


Fig. 3

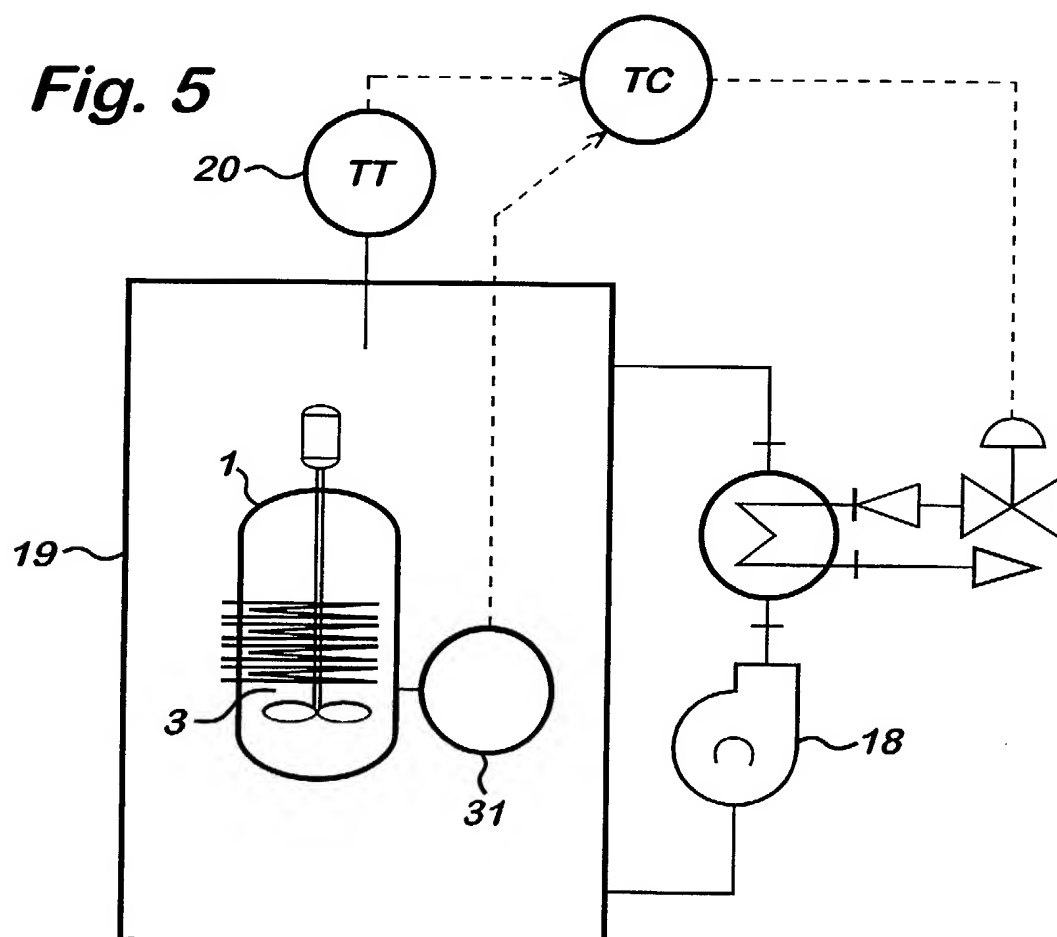
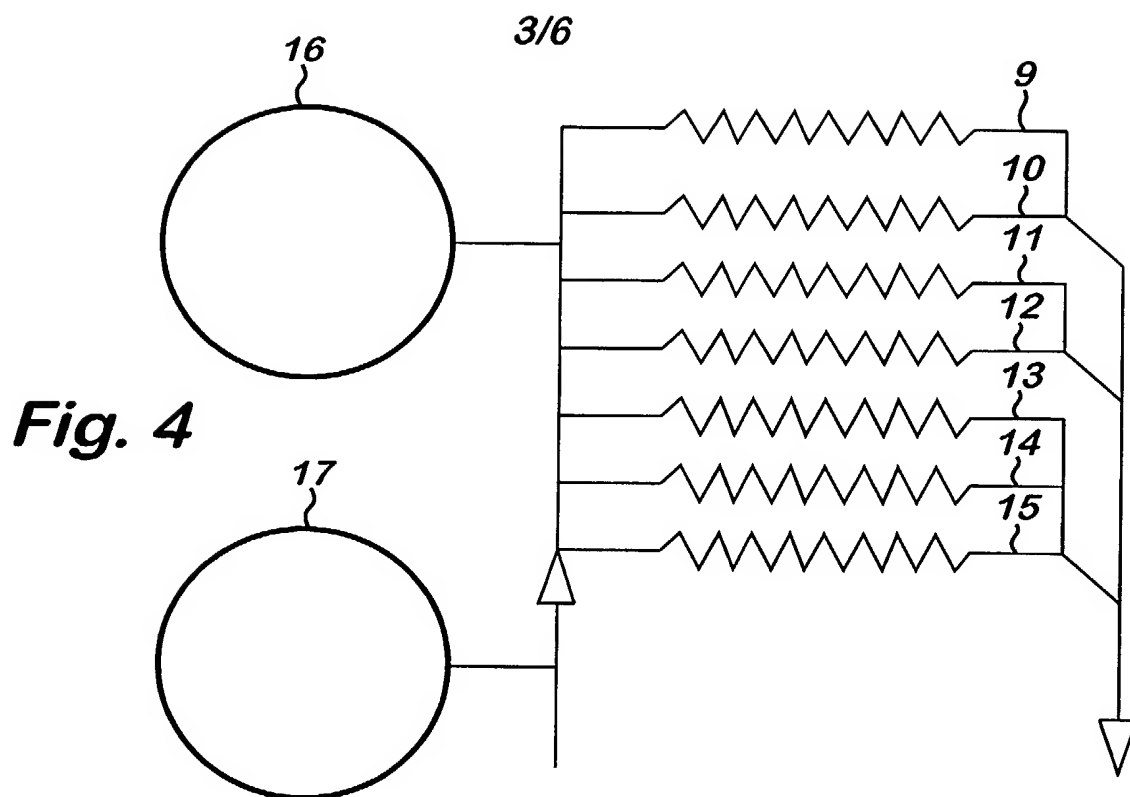


Fig. 6

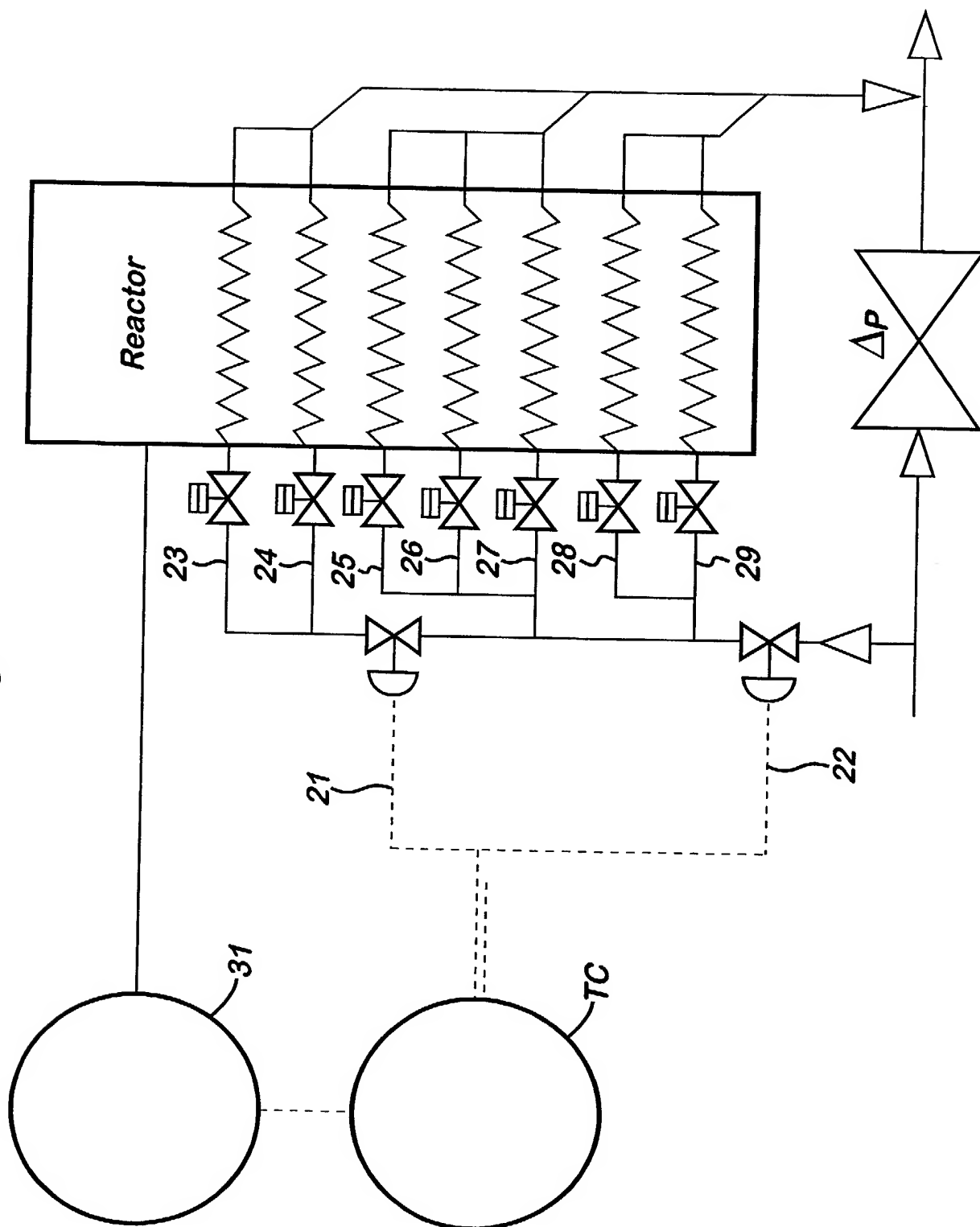
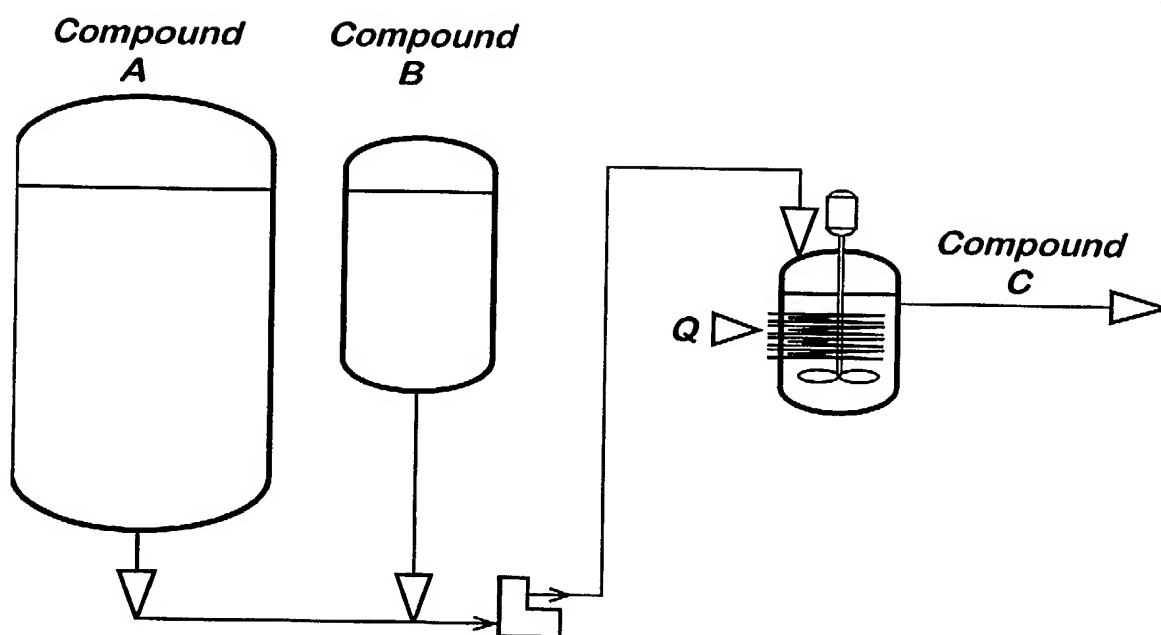
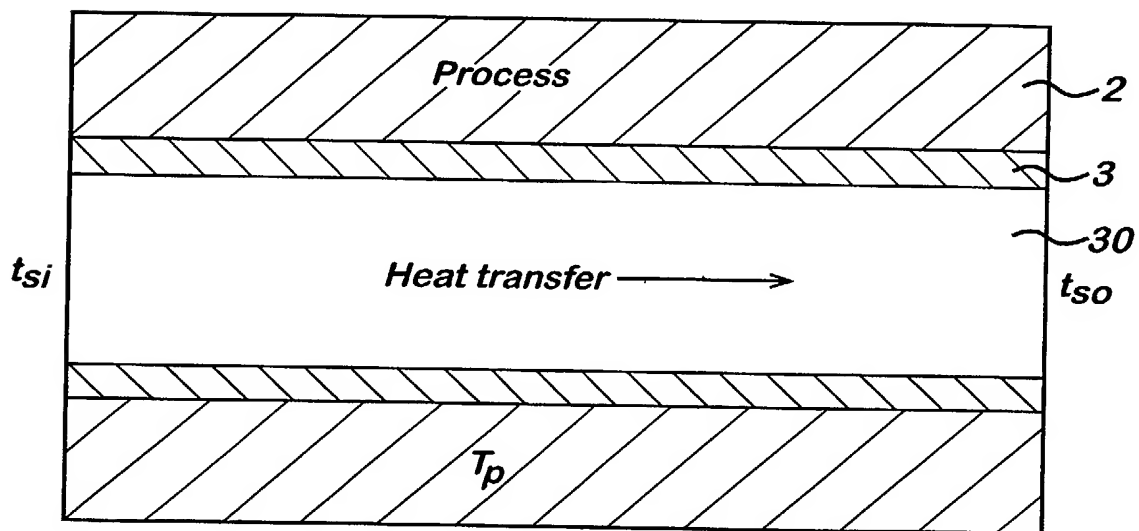


Fig. 7**Fig. 8**

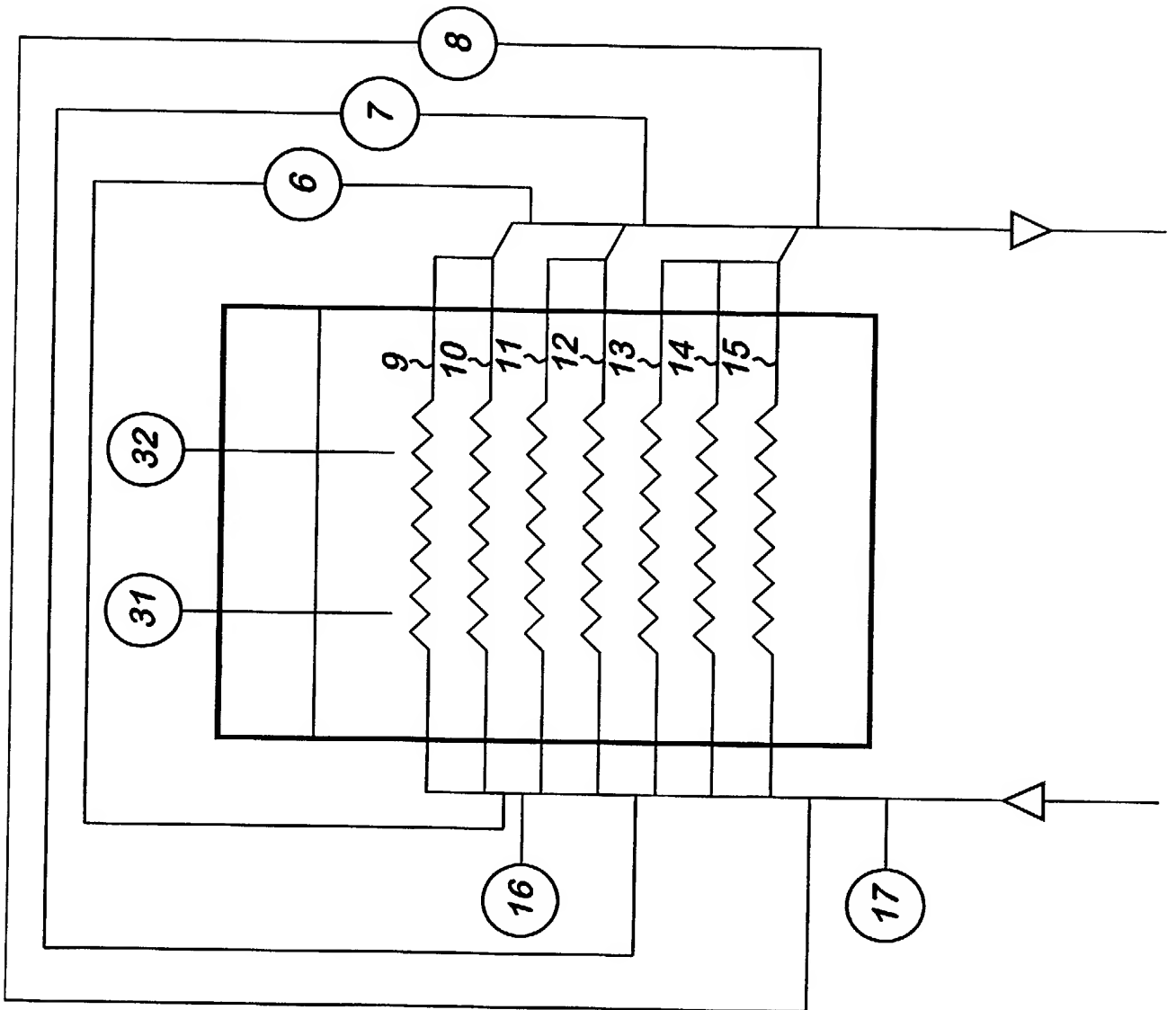


Fig. 9

IMPROVED REACTOR HEAT TRANSFER SYSTEMS

5 The present invention relates to reaction systems and in particular reaction systems
embodying improved heat transfer. Reaction systems may involve physical and/or
chemical changes. Chemical reactions involve chemical change such as in the reaction
of two or more molecules to produce a new molecule including polymerisation, in the
breakdown of molecules into two or more molecules. Physical reactions involve a
change of state such as in crystallisation, precipitation, evaporation, melting,
solidification and the like. Certain reactions can involve both chemical and physical
10 change.

The invention is concerned with improving the ability to monitor the progress of physical
and/or chemical reactions, it is also concerned with improving the control of reaction
systems through the improved monitoring. The improved control that is provided by the
15 present invention enables the production of materials of higher quality and purity, it
enables more efficient use of reaction equipment and can further improve the efficiency
of the equipment so that shorter reaction times are needed to obtain a given amount of
material from a given amount of starting materials. Another advantage is that smaller
reactors may be used to produce a given volume of material.

20 Many reactions are hazardous and care needs to be taken to ensure no accidents. The
more accurate and more timely monitoring of the reaction provided by this invention
enables reactions to be performed within stricter limits. This enhances safety and can
reduce the reaction inefficiencies that, hitherto, were an inherent shortcoming of the
25 manufacturing process.

Reactions whether they be physical, chemical or both generate or absorb heat and there
is therefore a heat change across the reaction. The theoretical heat generated or
absorbed in a particular reaction is known from established information. The actual heat
30 generated or absorbed during the course of a reaction could therefore, in theory, be a
useful measure to determine reaction efficiency in the case of steady state reactions and
reaction progress in the case of batch reactions.

By way of an illustration of the theory, a typical chemical synthesis step will be considered. Two reagents (A and B) react together to form a new compound (C) as follows:



5

where $A = \text{kmol of A}$

$B = \text{kmol of B}$

$C = \text{kmol of C}$

The heat generated by this reaction is established according to the formula:

10

$$Q = \Delta H_r \cdot C \text{ (kJ).}$$

where $\Delta H_r = \text{heat of reaction per kmol of C produced (kJ/mol)}$

$C = \text{kmol of component C produced (kmol)}$

15 The value of ΔH_r may be determined from theoretical data or laboratory calorimeters.

Currently the heat data described may be used in a variety of ways.

For any reaction, the maximum theoretical heat liberation can be calculated as follows:

20

$$Q' = \Delta H_r C' \text{ (kJ)}$$

where $Q' = \text{maximum theoretical heat generated (kJ)}$

$\Delta H_r = \text{heat of reaction per kmol of C produced (kJ/kmol)}$

$C' = \text{maximum theoretical yield of component C (kmol)}$

The maximum theoretical yield C' is based on the assumption that one or both of the
25 feed components (A and B) are completely consumed.

If the heat of reaction is measured during a process, the quantity of component C synthesised at any time is as follows:

$$C = Q/\Delta H_r \text{ (kmol)}$$

30

where $C = \text{quantity of C produced (kmol)}$

$Q = \text{heat measured during the reaction (kJ)}$

$\Delta H_r = \text{heat of reaction per kmol of C produced (kJ/kmol)}$

Thus the total mass of C can be calculated by knowing the total heat absorbed or liberated and the heat of reaction (or crystallisation etc).

5 The expected theoretical yield of C is known from the quantity of reactants present and the stoichiometry of the process. Thus from the information above, the percentage conversion can be determined from the equation below.

$$\eta = C/C' \times 100\%$$

10 Where η = percent conversion

C = quantity of C produced (kmol)

C' = maximum theoretical yield of component C (kmol)

15 In batch reactions, percent conversion (η) provides an effective means of identifying reaction end point. This can be used to reduce manufacturing time and improve plant utilisation.

20 In continuous (plug) flow reactors, reaction efficiency (η) provides a parameter for controlling feed rate to the reactor and controlling process conditions. In this way it is possible to run conventional batch processes in small-scale plug flow reactors. This benefits all aspects of the manufacturing process including lower capital cost for equipment, increased plant versatility, improved product yield, safer process conditions (through smaller inventories), greater product throughput and reduced product development time.

25 The ability to monitor reaction progress has an additional safety benefit for both small and large reactors. A system with online calorimetric data can instantly identify when unreacted compound is accumulating in the reactor. This reduces the risk of runaways due to accumulation of unreacted chemicals.

30 The design of reactors in common industrial use is however inherently unsuitable for measuring calorimetric data and thus the techniques described remain theoretical.

Chemical reactors in common use in, for example, the pharmaceutical and fine chemical industries fall into four main categories. Standard batch reactors in which reagents are mixed in a stirred vessel in which heat is added or removed by means of heat transfer fluid recirculating through an external jacket. These are the most commonly used reactors for small-scale organic and inorganic synthesis reactions. Batch reactors with internal coils, which are a variation on the standard batch reactor and have additional heat transfer surfaces within the body of the liquid. These reactors are used for general-purpose batch reactions where higher heat loads are encountered. Loop reactors in which reactants are pumped through an external heat exchanger and returned to the vessel. These are commonly used for gas/liquid reactions in which case the liquid is returned to the reactor via a spray nozzle to create a high gas/liquid interfacial area. Continuous reactors in which reactants are pumped through a heat exchanger under steady state conditions. These are generally used for larger scale manufacturing processes with long product runs.

The heat transfer characteristics of the four types of reactors described above have three common features:

- i. The heat transfer fluid is circulated through the heat exchangers at high velocity to maintain favourable heat transfer coefficients. In the case of jacketed reactors, this is achieved by injecting the heat transfer fluid into the jacket at high velocities using nozzles or diverting flow around the jacket with baffles. In some instances, coils for the flow of heat transfer fluid are welded to the outside wall of the reactor vessel.
- ii. High mass flow rates of heat transfer fluid are employed to maintain a good average temperature difference between the heat transfer fluid and the process fluid.
- iii. The heat transfer area is fixed and temperature control of the process fluid is achieved by varying the temperature of the heat transfer fluid. In some cases limited scope exists for increasing or decreasing the heat transfer area.

The features described above represent good design practice for achieving a flexible and optimised heat transfer capability within the reactor. However, these features do not lend themselves to measuring the quantity of heat generated or liberated. This deficiency is illustrated by reference to the chemical reaction between reagents A and B as discussed above. (It should be noted that the example is not limited to chemical reactions and is equally applicable to other chemical and physical processes).

When the two reagents (A and B) react together to form C, heat is liberated. The heat liberated per second can be expressed as follows:

$$q = \Delta H_r \cdot c \text{ (kW)}$$

where q = heat liberated per second (kW)

ΔH_r = heat of reaction per kmol of C produced (kJ/kmol)

c = kmols of component C produced per sec (kmol/s)

If the process temperature remains constant the heat liberated (q) will be observed as a temperature rise in the heat transfer fluid according to the formula.

$$q = m \cdot C_p (t_{si} - t_{so})$$

where q = heat liberated by the reaction (kW)

m = mass flow rate of the heat transfer fluid (kg/s)

C_p = specific heat of heat transfer fluid (kJ.kg⁻¹K⁻¹)

t_{si} = temperature of heat transfer fluid in (°C)

t_{so} = temperature of heat transfer fluid out (°C)

However, in order to determine q , the flow rate and temperature change of the heat transfer fluid ($t_{si} - t_{so}$) must be measured accurately. In the reactor examples described above, effective design favours high flow rates of heat transfer fluid. Often this leads to a temperature change of the heat transfer fluid ($t_{si} - t_{so}$) of less than 1°C. An IEC Class A RTD is one of the more accurate temperature measurement devices available. These devices have a tolerance of $\pm 0.25^\circ\text{C}$ (the error on the installed device may be higher). Thus for a temperature change of 1°C, the accuracy of heat measurement can be expected to be $\pm 25\%$ or worse. This would rise to 250% where the heat transfer fluid temperature changed by 0.1°C. This factor alone makes it virtually impossible to measure the heat of reaction in conventional reactors. Furthermore, on a conventional

reactor, heat leaking out of the system via the non-process side of the jacket can create serious error.

5 Furthermore, conventional chemical reactors often have sluggish control systems which permit temperatures of the bulk material to cycle by a few degrees. In energy terms a few degrees change in temperature can represent a significant proportion of the overall energy release.

10 Conventional reactors offer acceptable heat transfer characteristics when the flow of heat transfer fluid is held at a good velocity. Since the heat transfer surface is limited to 1 or 2 discrete elements, the range (of energy liberated or absorbed) over which a useful service temperature rise ($t_{si}-t_{so}$) can be achieved is very limited. In a case where the energy release from the process is small, the temperature rise in the heat transfer fluid may be a fraction of a degree. In addition to this, the shaft energy of the heat transfer
15 pump could be a high proportion of the total.

The limitations described above are common to all reactors (and evaporators, batch stills etc) used in the pharmaceutical, chemical and allied industries. Accordingly, when employing these reactors the heat generated or consumed by the reaction cannot be
20 used to monitor the progress of a reaction within any degree of accuracy.

It has been proposed in US Patent 6106785 that the heat generated in a polymerisation reaction may be used to monitor the progress of the reaction. The system of US Patent 6106785 is however a coarse method for monitoring a reaction which involves employing
25 an inferential sensor, whose concept is based on the observation that for polymerisation processes, the amount of heat released is proportional, albeit in a non-linear way, to the degree of the monomer conversion. Accordingly to US Patent 6106785 by careful calculation of the reactor's thermal balance on-line one can continuously infer the degree of conversion and use it for control. Once the actual degree of conversion can be
30 determined and ultimately controlled, one can also control the cooling duty of the reactor and thus make it conform with the cooling capacity allotted to it by the plant scheduler. US Patent 6106785 is therefore concerned with optimising the use of heat transfer fluid and the addition of initiator/inhibitor within safe operating parameters.

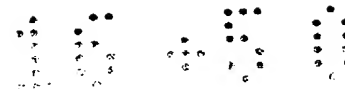
In US Patent 6106785 the batch controller data is used directly to control the reactor mixture temperature by manipulating the incoming coolant flow and temperature. The data are fed into the inferential sensor, where they are used to infer the current value of the degree of monomer conversion.

5

In US Patent 6106785 the degree of conversion is not therefore measured directly, but it is inferred by dynamically evaluating the reactor heat balance. US Patent 6106785 therefore enables one to infer the degree of conversion from the dynamic evaluation of the reactor heat balance. The use of the degree of conversion replaces special sensors for feedback control with respect to the product quality (end-use) properties. The use of the degree of conversion also replaces physical time for the timing of process related operations like valve opening and closing, and enables control of the heat supply/removal, dosing of the reactants, and so forth. The use of the sensor is said to allow an increase in the accuracy of the prediction of the batch evolution and thus enables a more accurate prediction of the cooling need profile than that provided by the systems previously used.

In US Patent 6106785, the reaction mixture temperature and the integral heat rate are treated as two independent process variables. This approach is said to allow the user the freedom to specify batch recipes in a way that defines the evolutions of either variable during the batch run, and to execute them under tight, high performance control. Because the degree of monomer conversion is proportional to the integral heat rate for many important polymers including PVC, controlling the two variables is said to allow the user independent control over two basic determinants of products quality. According to US Patent 6106785 this control fully defines the heat release at every instant of the batch run, thus making it possible to better utilize the available cooling capacity through more reliable planning and scheduling. To control the temperature and integral heat rate independently, the proposed method manipulates the amount of heat added to or taken out of the reaction and the amounts of the initiator(s) and inhibitor added during the batch run.

Whilst these techniques bring benefits in optimising the use of the coolant they are not sufficiently accurate and discerning to enable sophisticated sensing and control of a



reaction. They are also not applicable to a reactor operating over a wide range of conditions. The present invention provides the solution to this problem.

In our co-pending Application reference PAAMBA092 filed on the same day as this Application we describe and claim reactor systems which provide improved control over physical and/or chemical reactions. The present invention relates to PAAMBA092 in that it enables the improved control to be achieved over a wide range of operating conditions by the use of a variable heat transfer area between the process fluid and the heat transfer fluid. Our co-pending Application PAAMBA094 also filed on the same day as this Application describes measurement systems which may be used with this invention.

Accordingly the present invention provides a reaction system in which the heat transfer fluid is carried through a process fluid in a conduit and the heat transfer surface area between the conduit and the process fluid can be varied according to the heat generated or absorbed by the reaction as determined by the temperature change in the heat transfer fluid across the reaction and the mass flow of the heat transfer fluid through the conduit.

Whilst any form of conduit may be used for the heat transfer fluid, pipes and coils are preferred and the invention will hereafter be described in relation to the use of a coil or coils.

In this invention the heat transfer fluid serves as both process temperature controller and as a heat flow measuring device. In order for effective performance of the invention the following operating conditions are preferred.

- i. the average temperature difference between the heat transfer fluid and the processes fluid is from 1 to 100°C
- ii. the temperature differential ($t_{sf}-t_{so}$) of the heat transfer fluid across the reaction system is at least 1°C
- iii. the linear velocity of the heat transfer fluid is at least 0.1 meters/second.

We have found that providing these criteria are satisfied measurement of the flow rate and temperature change of the heat transfer fluid across the reaction enables the heat

generated or absorbed by the reaction system to be determined with a high degree of accuracy. The determination may then be used to optimise the surface area of the conduit (coil) for the heat transfer fluid that is made available to the process fluid which in turn enables one to monitor and control the reaction with a high degree of accuracy over a wide range of reactor operating conditions.

In order for effective operation, the reaction system should preferably have the following characteristics:

- 10 a. The available surface area of the heat exchanger should be sufficient to ensure that a measurable temperature difference ($t_{si}-t_{so}$) is observed in the heat transfer fluid as it passes across the reactor. For the purposes of accuracy, a temperature difference of more than 1°C (preferably more than 5°C, more preferably more than 10°C) is desirable.
- 15 b. A high temperature difference is preferably maintained between the process fluid and the inlet heat transfer fluid (t_{si}) to ensure that an accurately measurable service fluid temperature change ($t_{si}-t_{so}$) can be achieved and smaller heat transfer areas are required.
- c. As far as possible, heat must only be transferred to or from the process fluid and not be transferred to other equipment or the environment.
- 20 d. The heat transfer fluid must always flow at a reasonable velocity. The velocity will vary with coil size and conditions but it is preferred that it is greater than 0.1 m/s more preferably greater than 1 m/s. Lower velocities will give slower temperature control response. Low velocities also give a higher ratio of thermal capacity (of the heat transfer fluid) to heat release rate. This will compound errors in the values of measured heats.
- 25 e. When used for batch processes or multi-purpose duties, the heat transfer equipment should be capable of stable operation over a wide range of energy release/absorption rates. The range will vary according to the nature of the reaction. In the case of batch reactions the ability to vary the available surface area of the heat exchanger enables operation over a very wide range.
- 30

To satisfy condition c above, the variable area heat exchanger is preferably immersed in the process fluid and should be fully insulated at all points other than where fully

immersed in the process fluid. This ensures that all the heat gained or lost by the heat transfer fluid is transferred directly from and to the process fluid. This condition is most easily achieved by designing the heat exchanger as a coil or a series of coils fully immersed in the process fluid.

5

It is further preferred that an optimal relationship between heat transfer surface area to heat transfer fluid flow capacity is provided. Such conditions exist when the heat transfer fluid (traveling at the desired linear velocity) provides an easily measured temperature change (such as 10°C) without incurring excessive pressure drop. It should be noted that the optimum heat transfer conditions vary according to the properties of the process fluids and heat transfer fluids respectively.

10

In order to satisfy these criteria, the heat exchanger for the reactor is preferably a series of heat transfer coils, which pass through the reaction fluid, the coils may be brought in and out of action to vary the available heat transfer area according to the needs of the reaction. The design of the coils is important to achieving the object of the invention and must be such that the heat transfer area matches the heat carrying capacity under specified conditions.

15

The techniques of the present invention may be used in systems in which the heat transfer fluid is straight through or recycled.

20

The heat transfer area of a coil may be related to the flow carrying capacity of the liquid by using the formula

25
$$U.A.LMTD = m.Cp.(t_{si}-t_{so}) \text{ (kW)}$$

where U = overall heat transfer coefficient ($\text{kW.m}^{-2}.\text{K}^{-1}$)

A = heat transfer area (m^2)

m = mass flow rate of heat transfer fluid (kg/s)

30 $LMTD$ = log mean thermal difference between service and process fluids ($^{\circ}\text{C}$)

Cp = specific heat of heat transfer fluid ($\text{kJ.kg}^{-1}\text{K}^{-1}$)

$(t_{si}-t_{so})$ = temperature ($^{\circ}\text{C}$) change in the heat transfer fluid between inlet and outlet

This information may then be used to determine the optimum diameter to length relationship of an individual coil whereby high turbulence is achieved without incurring excessive pressure drop through the heat exchanger:

- 5 a. The temperature difference between the inlet heat transfer fluid and the process fluid should be large enough (e.g. 5-100°C) to ensure that the heat transfer fluid undergoes a measurable temperature change ($>1^{\circ}\text{C}$ or preferably greater than 10°C) in its passage through the coil. The temperature change must not however be so high or low as to cause freezing, waxing out, boiling or burning of the process fluid.
- 10 b. The heat transfer area must be large enough to ensure that the heat transfer fluid undergoes a measurable temperature change ($>1^{\circ}\text{C}$ or preferably greater than 10°C) through the process fluid. Smaller temperature changes limit heat transfer capacity and accuracy. Higher temperature changes are desirable providing they do not cause freezing, waxing out, boiling or burning of the process fluid.
- 15 c. The linear velocity of heat transfer fluid must be reasonably high (preferably $>0.1 \text{ m.s}^{-1}$) in order to maintain satisfactory control response and a good overall heat transfer coefficient.
- d. The pressure drop of heat transfer fluid flowing through the coil is from 0.1 to 20 bar.
- 20 In practice, optimum coil lengths will vary according to the temperature differences employed and the thermodynamic and physical characteristics of the system. Calculating the optimal length of each coil is an iterative process. A general-purpose device will be sized using conservative data based on fluids with low thermal conductivity and a low temperature difference between the reaction fluid and the heat transfer fluid.
- 25 Each coil will have a limited operating range.

In the system of the present invention in which the heat transfer equipment is capable of stable operation over a wide range of energy releases, the system is such that the area of heat transfer may be varied according to the needs of the particular reaction (or stage of reaction). This may be conveniently accomplished by providing multiple heat transfer coils each of which has a diameter and length relationship designed to provide a certain degree of heat transfer. In this preferred multiple coil system, the coils may be brought into and out of operation as the needs of the reaction system dictates.

The reaction system of the present invention is described with reference to the accompanying drawings in which Figure 1 is a schematic illustration of a reaction vessel served with a single heat transfer coil (of specified diameter) and is included to illustrate the sizing of individual coils which has applicability to the multiple coil systems of this invention. Figure 2 is a schematic illustration of a comparable reactor served with three heat transfer coils to provide a variable heat transfer surface according to the invention.

Figure 1 is a schematic illustration of a reactor (1) containing a process fluid (2) and a cooling coil (3) which is three meters long. This system is capable of accurately measuring energy changes of between 72 and 260 watts. Measuring energy release rates of less than 72 watts is achieved at the expense of lower accuracy (smaller temperature rise in the heat transfer fluid) or slower control response (slower velocity of heat transfer fluid). Measuring energy release rates of greater than 260 watts, introduces the risk of freezing (or burning/boiling where heat was being applied) as very cold (or hot) heat transfer fluid has to be supplied. The alternative of higher heat transfer fluid flow delivers only moderate improvements (slightly improved U value and higher temperature difference between process and service fluids) in terms of heat transfer capacity and is achieved at the expense of progressively lower accuracy (smaller temperature rise in the heat transfer fluid).

The reactor in Figure 2 overcomes these problems and has an improved measuring range of 72 to 780 watts. The versatility has been increased by adding two more coils (4) and (5). When one coil is operating heat generation in the range of 72 to 260 watts can be measured (as in the reactor of Figure 1). With all three coils operating (at a nominal maximum flow) up to 780 watts can be measured. By this method, it is possible to design a reactor with a wider operating range.

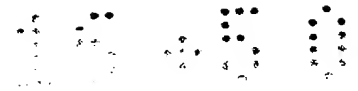
In normal operation, the flow of heat transfer fluid to a set of coils will be increased using a flow control valve. When a new coil switches in to accommodate a rising load, the control valve will regulate the flow to ensure smooth transition to the higher flow. This will require a rapid flow control response to the step change in the system pressure drop. To provide a smooth transition between operating conditions and wide operating range a large number of coils is desirable.

Instrumentation is a key aspect of successful operation of the systems of the present invention. Accurate and sensitive instrumentation must be used for measuring temperatures, the rate of flow of the heat transfer fluid and increasing or reducing the heat transfer area. Instruments must operate over a wide range of flows and this may be achieved by breaking up the coil system into separate modules operated by manifolds. This enables different coils to be brought into or out of operation according to the needs of the system.

Fast and accurate temperature measurements is a key performance requirement. To achieve this, the temperature element is conveniently mounted in fast flowing heat transfer fluid. A minimum hold up volume (of service liquid) should exist between the temperature elements and the heat transfer surface. This is achieved by using sub manifolds on the discharge pipes as shown in Figure 3.

Figure 3 is a schematic illustration showing three differential temperature measuring devices (6), (7) and (8) on a seven-coil system based on coils (9) to (15). These devices measure temperature change of heat transfer fluid flowing across the coils. The temperature devices work in a cascade fashion. At low flow (coil 9 or coils 9 and 10 operating) measuring device (6) is used for measuring discharge temperature. When three or more coils are operating, measuring device (6) switches to idle and measuring device (7) takes over. When five or more coils are operating, both (6) and (7) switch to idle and measuring device (8) takes over. This concept is applied irrespective of the number of coils and temperature devices used. It is preferred that the linear velocity of the heat transfer fluid as it passes the temperature element is one meter per second or greater (although slower velocities can be tolerated). The temperature devices must be highly accurate and sensitive. It should be noted that separate inlet and outlet temperature devices could be used as an alternative to the differential devices.

In a preferred process, in addition to the normal process temperature transmitters, which constantly measure the process across its entire range and provide the necessary safety interlocks, a second pair of temperature elements can be provided to monitor the specific process set point. This arrangement uses two different types of measuring elements. The main device is preferably an RTD, a 4 wire Pt100 RTD to 1/10th DIN standard being especially suitable. The transmitter used to provide the 4-20 mA output signal is



spanned to the minimum allowable for the transmitter (similarly any output signal type or temperature span could be used). The temperature transmitter will be calibrated specifically at the process set point. Larger ranges will still give acceptable results, but reducing the span to the minimum possible offers improved accuracy and resolution.

5 Thus this arrangement will provide an extremely accurate means of process temperature measurement.

The element of the temperature measurement system is the part of the device which is in contact with the liquid. In the case of an RTD, its resistance will change in response

10 to changing temperature. The response of an RTD is not linear. The transmitter is the calibrated part of a measuring device and is used to linearise the output to the control system and convert the signal to an industry standard, usually 4-20 mA, but it could also be 1-5 V or 0-10V. A thermocouple's response to a change in temperature is a varying voltage. Usually milli volts per °C. A thermocouple transmitter will again convert this

15 signal to an industry standard, again more often than not, 4-20 mA. Accordingly the term 'element' is used when describing a physical presence in the process, e.g., a temperature element is located in the reactor and measures the temperature of the reactor contents. The term 'transmitter' when describing aspects of temperature measurement relating to the control system, e.g., a temperature transmitter is calibrated

20 0-100 °C and displays the contents temperature of the reactor.

The limitation of any RTD is its speed of response to a step change in temperature. Typically it can take up to four or five seconds for an RTD to measure a change in temperature. Thermocouples, on the other hand, can respond much more rapidly to

25 temperature fluctuations. For this reason a thermocouple is also used to monitor the process set point, a T type thermocouple being especially suited. Its transmitter will be similarly ranged to the RTD. However, as a T type thermocouple has an accuracy of only + or - 1°C, it will not be used to monitor the process temperature. Its function is to monitor the rate of change of the process temperature.

30 The combined use of these two different types of sensing elements provides a temperature control system, which is both extremely accurate and responsive. It should be noted that not all process operations require this level of temperature measurement

accuracy and control. In such cases, more basic temperature control and measurement systems will prove tolerable.

5 In order to fully utilize this two-element approach, custom software is used to determine which process variable (temperature, or rate of change of temperature) is the most significant at any one instance in time.

10 Accurate measurement of flow is also important for effective operation. Figure 4 shows a flow measurement system for the reactor shown in Figure 3 employing multiple flow devices. Flow device (16) is a low range device for measuring flow when coils (9) or coils (9) and (10) are in operation. When three or more coils are in operation, flow device (17) takes over and (16) switches to idle. Any number of flow transmitters can be used to achieve satisfactory accuracy. As a general rule, the number of flow devices to be used should be calculated as follows

15
$$\text{Number of flow devices} = (F_{\max} - F_{\min}) / (R \cdot F_{\min})$$

where F_{\max} = maximum flow (kg.s^{-1})

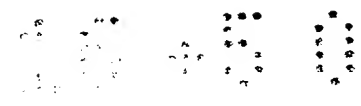
F_{\min} = minimum flow (kg.s^{-1})

R = turn down ratio of the flow instrument

20 The above equation makes reference to mass flow. The equipment can use a volume flow device however provided the system converts volume flow data into mass flow data. This can be done automatically by the control software (mass flow = volume flow x liquid density). For sensitive systems (or those with a wide temperature range) compensation
25 should be made for changes in liquid density. Information on liquid density can be input manually into the control system. Alternatively, the control software can calculate the density based on temperature using established mathematical relationships.

30 In the present invention, the reactor is operated at constant temperature. Any losses or gains in temperature to the environment will be recorded as reaction activity. In our preferred system three measures are used to take this into account.

- Heat losses are compensated for by zero calibration prior to reaction
- The vessel is lagged or located in a box to minimise heat loss



- For very sensitive systems the insulating box is temperature controlled by an independent loop as shown in Figure 5.

The arrangement in Figure 5 shows a second heating cooling loop with a fan (18) circulating air within the temperature-controlled box (19). The air temperature within the insulated box is determined by temperature measurement device (20) and is maintained at the process reaction temperature (31). This eliminates heat loss/gain to/from the environment.

- 10 Any net heat flows in and out of the system must be monitored or controlled. Where liquids (or dry gases) enter or leave the system, these should be at reaction temperature. If not they should be of known specific heat and monitored for temperature and flow. Vapour carried out of the system presents a greater problem. If the gas flow is significant, two options can be employed. Heat losses with off gas are measured in trial runs and compensated for in the calculations. This solution has to be used with care for batch operation when gas evolution varies with time. Accordingly for batch operation it is preferred that the gas flow out of the reactor be measured and the information translated into heat flow data.
- 15
- 20 The system works most effectively under isothermal conditions. It can however be used for reactions where the process temperature changes. In this case it is necessary to measure the heat capacity of the system as follows:

$$\Sigma M.C_p = (M_p.C_{p_p}) + (M_c.C_{p_c})$$

25

Where $\Sigma M.C_p$ = heat capacity of the system (kJ/°C)

M_p = mass of process fluid (kg)

C_{p_p} = specific heat of process fluid (kJ.kg⁻¹K⁻¹)

M_c = mass of equipment in contact with process fluid (kg)

30

C_{p_c} = specific heat of equipment in contact with process fluid (kJ.kg⁻¹K⁻¹)

In practice $\Sigma M.C_p$ may be calculated by using the reactor. This is achieved by heating or cooling the process fluid and measuring heat lost or gained over a given temperature change when no heat is being absorbed or liberated by the process.

$$\Sigma M.C_p = Q/(t_s - t_f) \quad (\text{kJ}/^\circ\text{C})$$

Where $\Sigma M.C_p$ = specific heat of the system (kW/°C)

Q = measured quantity of heat added or removed (kJ)

5 t_s = temperature at the start of heating or cooling (°C)

t_f = temperature at the finish of heating or cooling (°C)

10 This heat capacity information may be fed into the system and used as a correction factor when the temperature changes during the process. The heat capacity information also serves as useful process data.

Conventional reactors have fixed area heat transfer surfaces (or occasionally several elements such as separate sections on the bottom dish and walls). They perform most effectively with a high and constant flow rate of heat transfer fluid to the jacket (or coils).
15 Process temperature is controlled by varying the heat transfer fluid temperature. In the system of the present invention, the area of the heat transfer surface is varied according to the needs of the reaction (although some variation in heat transfer fluid temperature can also be used).

20 A typical control arrangement for control of the heat transfer fluid using a variable area heat transfer surface according to this invention is shown in Figure 6. In Figure 6 valves (21) and (22) are control valves that regulate flow of heat transfer fluid to the heat transfer coils. The extent to which they are open is determined by a temperature output measure from the reactor (or vessel). With the process at idle, valve (23) is open and
25 sufficient flow permitted to compensate for heat gain from the agitator. As load is applied to the process, valve (21) opens to permit the flow of more heat transfer fluid. When valve (21) is open beyond a pre-set point (or when flow rate dictates) valve (24) will open and valve (21) will close up slightly to compensate. As valve (21) approaches the top of its control range, valve (22) takes over. As valve (22) progressively opens the
30 valves (23) to (29) are opened in a cascade fashion.

The required number of flow control valves can be calculated in the same manner as for flow devices (see above).

Any number of control valves can be used and they can be installed in series (as shown) or parallel. In this preferred system the extent to which valve 21 and 22 are open is dictated by the process load. The number of on/off valves (which turn the coils on and off) open is dictated either by the position of the control valves or the measured flow.

5

The heat transfer fluid is applied to the control equipment at constant pressure and temperature. In some cases temperature can also be varied where it is necessary to increase the operating range.

10 A key requirement of this invention is reliability. This is particularly important in pharmaceutical applications where current good manufacturing practice (cGMP) dictates that the equipment operates within stated design parameters.

15 To provide a means of calibration and as a performance check, the reactor may be fitted with an electrical heater (or some other type of reference heater). By supplying a measured current to the heater, reliable reference loads are provided for calibrating the system and checking performance. In pharmaceutical applications, control and data acquisition systems together with software should be validated to comply with cGMP standards.

20

The equipment incorporates both conventional instrumentation and process specific instrumentation. These process specific instruments operate at a higher than normal accuracy when compared to conventional instrumentation. Figure 9 is a schematic illustration of typical process instrumentation which consists of:

25

- A process temperature RTD instrument (31)
- A process temperature thermocouple instrument (32)
- heat transfer fluid differential temperature instruments (6), (7) and (8)
- heat transfer fluid flow meter instruments (16) and (17)

30

For the process temperature RTD instrument (31) and the heat transfer fluid differential temperature instruments (6), (7) and (8), matching the RTD sensor to the temperature transmitter can result in significant improvements. The specific characteristic of an RTD sensor is unique to each device. By storing this information in the transmitter

improvements in accuracy are obtained. The constants used in this technique are known as the Callendar-Van Dusen (CVD) constants. The present invention is unique in that it uses additional calibration steps to enhance the accuracy of its instrumentation. For example, if the CVD technique is coupled with the use of high accuracy RTDs (typically class B to 1/10th DIN standard) process specific calibration may then be carried out to bring about further improvements in accuracy.

By 'process specific calibration', (e.g. the optimum reaction temperature) we mean that the instrument is calibrated specifically at the normal process set point of an instrument and that the measuring system error is adjusted, such that at this operating point best accuracy is achieved (for a normally calibrated instrument, best accuracy is usually given at the maximum calibrated range, or at a point dictated by the characteristics of the sensor). For example if a process is to be controlled at 35°C, instrument (31) would be calibrated across a small range, say 25 to 45°C. Furthermore, the instruments would be calibrated at 35°C and adjusted so that at this specific point the error of the measuring system is the minimum achievable. Once installed and connected to the control system, the calibration of the instrument loop can be verified as a complete installation and any control system errors compensated for. The control system hardware is designed to minimise errors (precision components must be used) and thus optimise accuracy. Similarly the instrumentation installation must be such as to minimise measuring error. The use of these additional steps, will allow maximum possible calibration accuracy to be obtained.

The process temperature thermocouple (32) will be calibrated in a similar manner, but as it is used to measure rate of change of temperature as opposed to temperature, its overall accuracy, although still important, is less significant.

The heat transfer fluid differential temperature measuring instruments (6), (7) and (8) will also employ this same technique to ensure best calibration accuracy is achieved.

For the heat transfer fluid flow instruments (16) and (17) the technique is again similar. Calibration in this instance is carried out over a small operating range with the emphasis on achieving the best accuracy at the preferred flow. By using multiple instruments calibrated over relatively small operating ranges, e.g. 0-1, 1-2, 2-3 etc., a significant

improvement in accuracy is achieved than by using a single instrument calibrated over the range 0-3. Best accuracy is achieved by using a suitably sized instrument with a normal flow of 80 to 90% of the instrument span. Again, once installed in the field and connected to the control system, the calibration of the instrument loop should be verified as a complete installation and any control system errors compensated for. The control system hardware is again designed to minimise errors and thus optimise accuracy.

Routine calibration of the heat measuring equipment may be carried out in several steps as follows:

10

The first step is zero calibration. For accurate operation, zero calibration should be carried out for each type of process used. This permits the control system to compensate for any 'non-process' energy changes (e.g. heat gains and losses to the environment, energy gain from the agitator etc). The vessel is filled with liquid and the agitator switched on. It is then heated to the reaction temperature. When the temperature is stable at the operating temperature, the heating/cooling system will function at a very low level to compensate for non-process energy changes. The control system is zeroed under these conditions.

15

20

The second stage is to range and span the system. This is carried out by heating or cooling with a reference heater cooler. This may be in the form of an electrical heater or an independent heating/cooling coil. Heating (or cooling) is carried out at several different energy input levels to range and span the system.

25

Alternatively the instruments may be tested individually in which case the second step of the above process may not be necessary.

30

We have found that the reactor systems of the present invention are extremely useful as batch chemical synthesis reactors. We have also found that the invention enables the same size of machine to be employed for development, pilot plant and full manufacturing purposes.

The variable area heat transfer reactor of this invention is ideal for fast exothermic reactions, where it can operate as a small continuous flow reactor on processes hitherto

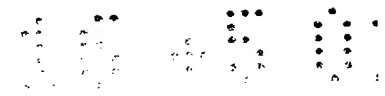
conducted as batch reactions. Unlike large conventional batch reactors, it is possible to operate in this mode as the reaction is continuously monitored. Any fall off in conversion efficiency is detected immediately and forward flow is stopped. The arrangement for this system is shown in Figure 7. The benefits of operating in this mode are various. The capital cost of a reactor for this type of application is substantially lower than a conventional reactor. In addition higher throughputs can be achieved. This type of equipment is also ideal for dangerous reactions as the inventory of reactants can be much smaller than that needed for conventional reactors. The equipment can also be programmed to stop reagent addition if unconsumed reactant starts to accumulate.

The invention is also useful in slow exothermic reactions even where large liquid volumes are held. In these reactors the data is obtained, analysed and used in a manner similar to the continuous reactor described above. The benefits of using this equipment for slow reactions is that the addition rate of the components can be regulated to prevent accumulation of unreacted chemicals. It is also possible to identify the end point of the reaction which offers substantial savings in plant utilisation as the product can be transferred forward with the confidence that it satisfies a key quality control objective. In some cases, accurate identification of end point also enhance product quality and yield.

The rate at which heat can be transferred between the process fluid and the heat transfer fluid is dictated (in part) by the overall heat transfer coefficient (U). The larger the value of U, the smaller the heat transfer area required. The U value may be calculated from three components.

- The heat transfer resistance through the process fluid boundary layer
- The heat transfer resistance through the coil wall
- The heat transfer resistance through the heat transfer fluid boundary layer

The boundary layers are the stagnant layers of liquid either side of the coil wall. The faster the agitation (or liquid flow), the thinner the boundary layer. Thus high flow rates give better heat transfer. Also liquids with good thermal conductivity give better heat transfer through the boundary layers.



Heat transfer mechanism across the coil wall is similar, except (unlike the boundary layers) the distance through which the heat has to conduct is fixed. Higher heat transfer rates are achieved where the coil material has high thermal conductivity. Higher heat transfer rates are also achieved where the coil material is thin.

5

Thus a high U value requires both a thin coil material (with high thermal conductivity) and turbulent conditions in both liquids (the more turbulent, the better). The higher the U value, the smaller the area required for heat transfer. This means a shorter heat transfer coil.

10

It is therefore preferred to use the thinnest walled coils possible without compromising mechanical strength and corrosion tolerance. A typical wall thickness would be $\frac{1}{2}$ to 4 mm.

15

The material from which the coils is fabricated is not critical but it should be inert to the process fluid. Preferred materials include stainless steel for non-corrosive organic fluids, Hastelloy C (22 or 276) or similar alloys for most reactions using chlorinated solvents or other corrosive compounds, tantalum and titanium would be used where special corrosive conditions existed. In some applications other materials such as plastic, glass, glass lined steel or ceramics could be used.

20

The techniques of the present invention can be used for measuring heat of physical changes such as the heat of crystallisation and evaporation.

25

For purposes of illustration only the following examples show the sizing of the heat transfer coils.

30

Example 1 illustrates the sizing of an individual heat transfer coil such as that used in the system of Figure 1. Examples 2 and 3 illustrate the sizing and use of multiple heat transfer coil systems which can be used to provide a variable heat transfer area according to this invention.

In these examples some of the numbers used are arbitrary and are chosen for purposes of illustration only. The examples illustrate the sizing of coils for a batch reactor where

an exothermic reaction takes place. In this, a theoretical reaction reagent *A* is reacted with reagent *B* to produce a new compound *C* as follows.



5 Where *A* = kg of *A*

B = kg of *B*

C = kg of *C*

The heat liberated ΔH_r is as follows:

$$\Delta H_{r_c} = 1,000 \text{ (kJ/kg}_c\text{)} \text{ (1)}$$

10

The batch reactor is prefilled with component *B*. Component *A* is added slowly (alternatively the two components could be pumped continuously through the reactor in the desired ratios). For the purposes of this example it is assumed that it is a fast reaction and component *B* reacts immediately on contact with *A*. The heat liberated is therefore proportional to the rate of addition (of *A*). If it is assumed that the addition rate is such that 0.001 kg/second of *C* is produced

15

The heat load of the reactor (*q*) = 0.001 x 1000 = 1 kW.

20

The reaction is also assumed to take place at constant temperature so that the heat load on the cooling fluid is also 1 kW.

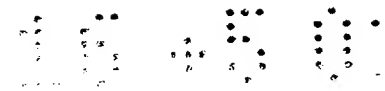
Figure 8 is a schematic illustration of a section through a typical heating/cooling coil such as coils (4) and (5) of Figure 2 in the process fluid (2) through which flows the heat transfer fluid (30).

25

Example 1.

The heat transfer coil (3) serves two functions, it controls the process temperature and also measures the quantity of heat liberated (or absorbed); for the purpose of this example, the term t_{si} is used for the measured inlet temperature of the heat transfer fluid and t_{so} for the outlet temperature of the heat transfer fluid. For effective operation, two factors need to be satisfied.

30



- i The temperature change in the heat transfer fluid ($t_{si}-t_{so}$) must be sufficiently large to provide a good measurable difference. For this example a 10°C temperature change of the heat transfer fluid ($t_{si}-t_{so}$) has been selected.
- ii In general, the temperature difference between the heat transfer fluid and the process fluid must be as high as possible but not so great that boiling, burning or freezing occur on the pipe surface. Assume that the reaction temperature is 30°C (t_p). Also assume that the lowest temperature at which service fluid can be delivered to the system is 5°C (to avoid freezing on the outer surface). Thus the service fluid inlet temperature (t_{si}) is 5°C and the outlet temperature (t_{so}) is 15°C [since ($t_{si}-t_{so}$) is 10°C].

Once the choice for ($t_{si}-t_{so}$) is made, the mass of the heat transfer fluid can be determined as follows:

$$m = q/C_p(t_{si}-t_{so}) \quad (1)$$

Where m = mass flow of heat transfer fluid (kg/s)
 q = heat gain by the heat transfer fluid = 1 (kW) (in this example 1 kW is the heat of reaction)
 C_p = specific heat of heat transfer fluid = $1.6 \text{ kJ.kg}^{-1}.\text{K}^{-1}$ (based on the choice of the synthetic heat transfer fluid)
 $t_{si} - t_{so}$ = temperature change of heat transfer fluid (selected to be 10°C)

Thus from equation (1), the mass flow (m) = $1/1.6 \times 10 = 0.0625 \text{ kg/s}$

Assume the density of the heat transfer fluid = 840 kg/m^3 .

Thus the volume flowrate of the fluid (W) = $0.0625/840 = 0.000074 \text{ m}^3/\text{s}$

Optimising coil geometry and the velocity of the heat transfer fluid is an iterative process. Low velocity of the heat transfer fluid through the heat exchange coil gives rise to poor control and measurement response. Low velocity also results in a large ratio of thermal mass of heat transfer fluid to heat load. This tends to magnify any errors of temperature measurement. High liquid velocity is desirable as it gives faster control response and a better ratio of thermal mass to heat load. As the velocity is increased however, the pressure drop through the coil gets higher.

Accordingly the optimum coil will be long enough to give adequate heat transfer area without incurring an excessive pressure drop. If the diameter is too small, the pressure drop will be too high (due to high liquid velocity and long pipe length). If the diameter is too large, the liquid velocity will be too low.

In this example an initial calculation based on a 4 mm diameter pipe is made for the first iteration as follows:

- At a flowrate of $0.000074 \text{ m}^3/\text{s}$ through a 4 mm bore pipe, the pressure drop of the heat transfer fluid is calculated as being 1.24 bar/m (based on synthetic heat transfer fluid).

The pipe length is calculated from the relationship

$$L = A / \pi D$$

- Where $L = \text{pipe length} = (\text{m})$
 $A = \text{surface area of pipe} (\text{m}^2)$
 $D = \text{pipe diameter} = 0.004 (\text{m})$
 $\pi = 3.1416$

- The surface area (A) required for control of the reaction is determined from the heat transfer capabilities of the pipe as follows:

$$A = q / U \cdot \text{LMTD} (\text{m}^2)$$

Where $A = \text{surface area of pipe} (\text{m}^2)$

- $U = \text{overall heat transfer coefficient} = 0.730 (\text{kW} \cdot \text{m}^{-2} \cdot \text{K}^{-1})$ (estimate for organic process fluid and synthetic oil heat transfer fluid)

$$\text{LMTD} = [(T_p - t_{si}) - (T_p - t_{so})] / \ln[(T_p - t_{si}) / (T_p - t_{so})] (\text{°C}) \text{ (log mean thermal difference between process and service fluids)}$$

$$\text{Also } T_p = 30$$

$$T_{si} = 5$$

$$T_{so} = 15$$

$$\text{Thus LMTD} = 19.6 (\text{°C})$$

$$\text{Therefore } A = 1 / (0.730 \times 19.6) = 0.07 \text{ m}^2$$

$$\text{Therefore } L = 0.07 / (3.1416 \times 0.004) = 5.6 (\text{m})$$

$$\text{The pressure drop through the line} = 5.6 \times 1.24 = 6.9 \text{ bar}$$

The linear velocity can also be calculated using the continuity equation as follows:

$$V = W/A$$

Where V = linear velocity (m/s)

W = volume flowrate (m^3/s)

5 A = cross sectional area of the pipe (m^2)

$$\text{Thus } V = 0.000074 / (\pi \times 0.004^2 / 4) = 5.9 \quad (\text{m/s})$$

A summary of the results of this calculation is shown in table 1 below.

Table 1

Coil duty	1 kW
Pipe diameter	4 mm
Liquid flowrate	0.074 l/s
Liquid velocity	5.9 m/s
Pipe length	5.6 m
Pressure drop	6.9 bar

10

The table shows that although the 4 mm diameter coil is capable of operating in a reaction that generates 1 kW of heat, it does so at the expense of very high pressure drop (of the heat transfer fluid). A small increase in process load beyond 1 kW would require even higher flowrates and a longer coil which would result in an unacceptably

15

high pressure drop. Thus under the conditions which have been chosen purely for the purposes of illustration, at a load of 1 kW the 4 mm diameter coil is at the top end of its operating range.

A larger pipe diameter of 5 mm internal bore is therefore selected for the second

20

At a flowrate of $0.000074 \text{ m}^3/\text{s}$ through a 5 mm bore pipe, the pressure drop of the heat transfer fluid is 0.42 bar/m (based on a standard pressure drop calculation synthetic heat transfer fluid).

25

The pipe length is again calculated from the relationship

$$L = A / \pi D$$

Where L = pipe length = (m)

A = surface area of pipe (m²)

D = pipe diameter = 0.005 (m)

$\pi = 3.1416$

- 5 The required area (A) is determined from the heat transfer capabilities of the pipe using the same formula

$$A = q/U.LMTD \text{ (m}^2\text{)}$$

as was used in the first iteration.

With the 5 mm coil however,

- 10 (note the value of U is lower in this case (0.66 kW.m⁻².K⁻¹) this is due to the reduced service fluid velocity (which gives a higher service side boundary layer resistance).

$$A = 1/(0.66 \times 19.6) = 0.077 \text{ m}^2$$

$$L = 0.077/(3.1416 \times 0.005) = 4.9 \text{ m}$$

The pressure drop through the line = 4.9 x 0.42 = 2.1 bar.

- 15 Also the new velocity is calculated as follows: Thus $V = 0.000074/(\pi \times 0.005^2/4) = 3.8$ (m/s)

The result of this second calculation are shown in table 2.

20

Table 2

Coil duty	1 kW
Pipe diameter	5 mm
Liquid flowrate	0.074 l/s
Liquid velocity	3.8 m/s
Pipe length	4.9 m
Pressure drop	2.1 bar

- 25 The 5 mm diameter coil therefore offers good linear velocities and a moderate pressure drop. Such a coil would therefore be useful for the operating conditions for the reaction used for the purposes of this example. The velocity is also well above the minimum preferred value (1 m/s).

To be of practical service, a heat transfer coil needs to operate over a range of conditions as opposed to being limited to one specific heat transfer rate. Table 3 shows

the performance of the 5 mm diameter coil under a variety of conditions (for organic process fluid and synthetic heat transfer oil). The one constant in the table is that the temperature change of the heat transfer fluid flowing through the coil ($t_{si}-t_{so}$) is always 10°C.

5

Table 3
CALCULATED COIL LENGTHS FOR A 5 mm ϕ COIL

Pressure Drop	Heat capacity	Flow	Velocity	LMTD 5°C	LMTD 10°C	LMTD 15°C	LMTD 20°C	LMTD 25°C
(bar/m)	(W)	(l/s)	m/s	(m)	(m)	(m)	(m)	(m)
0.1	457	0.033	1.7	8.9	4.4	2.9	2.2	1.8
0.25	761	0.055	2.8	12.4	6.2	4.2	3.0	2.5
0.50	1121	0.081	4.1	17.2	8.6	5.7	4.3	3.5
0.75	1439	0.104	5.3	20.8	10.4	6.9	5.2	4.2
1.00	1660	0.120	6.1	23.6	11.8	7.9	5.9	4.8

The first column in table 3 shows pressure drop (per metre of coil) through the coil for a given flow rate. The second column gives the heating or cooling capacity of the coil based on the 10°C temperature change. The third and fourth columns give the volume flow rate and velocity of the liquid. The last five columns give minimum coil lengths required for the quoted LMTD values. The LMTD temperature values quoted at the top of these columns represent the log mean temperature difference between the heat transfer fluid and the process fluid.

10

15

It can be seen from table 3 that different coil lengths are used depending on process heat load and log mean temperature difference between the process and service fluids. Table 3 shows that a large temperature difference is beneficial as it requires shorter coil lengths.

20

From table 3, a good general purpose coil for the reaction envisaged would be 5.9 metres in length. This would be capable of serving any of the duties contemplated in table 3 where the required coil length was 5.9 metres or less. It would be suitable for a process load of 1.66 kW providing the difference in temperature between process and heat transfer fluid was at least 20°C. Under these conditions the pressure drop through

25

the coil would be 5.9 bar. The coil also offers adequate heat transfer area and reasonable control response at heat loads down to 0.46 kW. Although low velocities are tolerable the control system becomes increasingly sluggish with low flows. Also low velocities result in a large ratio of thermal mass (of heat transfer fluid) to heat load. This
5 tends to magnify any errors of temperature measurement. High liquid velocity is therefore desirable as it gives faster control response and a better (lower) ratio of thermal mass (of the heat transfer fluid) to heat load.

For the reasons given above, high heat transfer fluid velocities are generally desirable.
10 Very high pressure drops however also introduce greater energy from turbulence and friction. There are also practical equipment constraints on how fast a liquid can be pumped through a pipe. The single coil system of example 1 is useful, but has its limitations.

15 As example 1 illustrates, a single coil has an optimum operating range. Although it is capable of measuring a range of heat transfer rates, it has its limitations. As table 3 shows, at heat transfer rates above 1121 W, the pressure drop across the coil increases rapidly due to the need for increasingly longer pipes and higher pressure drops per meter of pipe length.

20

The limitations of the single coil may be illustrated as follows:

A coil 6.2 m long operating with an LMTD (log mean temperature difference between the process fluid and service fluid) of 10°C has a nominal operating range of 457-1121 W. At
25 maximum load, the pressure drop across the coil would be 1.55 bar. If this coil was to be used with a heat load of 1660 W under the same conditions, it would have to be 11.8 meters long and the corresponding pressure drop would be 11.8 bar. If, under the same conditions, the LMTD was reduced to 5°C, the pipe would need to be 23.6 meters long and the resulting pressure drop would be 23.6 bar.

30

Although the range of a coil can be increased by varying the inlet temperature (t_{si}), there are limitations. If the temperature difference ($t_{si}-t_{so}$) is reduced, the system becomes progressively less accurate due to limitations of the temperature measuring devices. If the temperature difference ($t_{si}-t_{so}$) is expanded too far, there is a risk of freezing the

process fluid (or surface boiling or heat damage where heat is being absorbed by the process fluid).

5 Although service fluid flow and supply temperatures are both parameters that can be varied to alter the operating range, reliable control methods favour using one control parameter at a time (and step changing the other where necessary).

10 The 5 mm diameter coil illustrated in example 1 gives a turn down ratio of approximately 2.5 (1121/457). If the temperature difference across the coil ($t_{si}-t_{so}$) was increased from 10°C to 20°C, the turn down ratio could be increased to 5.

15 A method of increasing the operating range of the system is to employ variable area heat transfer according to the present invention and use multiple coils in a cascade fashion. Such a system is illustrated in example 2 in which the principles of coil sizing illustrated in example 1 are applied to a series of coils.

Example 2.

20 Example 2 illustrates, the design of variable area heat transfer systems employing multiple coil systems such as those illustrated in Figures 2 and 3. As in example 1, the cooling (or heating) coil system controls the process temperature and continuously measures the heat gained or lost using information on mass flowrate through the coil, temperature change ($t_{si}-t_{so}$) and specific heat of the heat transfer fluid.

25 Example 2 addresses the fact that a reactor might be required to handle exothermic reactions which generate heat in the range of 500 to 15,000 W. A range of this size exceeds the operating capabilities of the single heat transfer coil system illustrated in example 1. Such a reactor can however be effectively operated using multiple coils as illustrated in this example (in this example identical coils each 11.8 m long are used) in a cascade fashion. With one coil operating with the heat transfer fluid at 1.7 m/s, a heat
30 load of 457 W will give a temperature rise in service fluid ($t_{si}-t_{so}$) of 10°C. If, under the same conditions the velocity of the heat transfer fluid is increased to 6.1 m/second the capacity rises to 1,660 W (see table 3). If two coils are used at maximum flow the capacity is 3,320 W. By adding coils in this manner ever greater heat loads can be measured. If, for example, ten coils are used at the maximum flow, the capacity is

16,600 W. This system therefore offers a turndown ratio of approximately 36 (16,600/457). Accordingly, by varying the velocity of the fluid and the number of coils, the heat capacity can be measured with a high degree of accuracy over a wide range.

5 The devices described so far have turndown capacities of up to 36. In practice, a turndown of 1000 or more may be desirable. This could be important with a batch reaction where the end point needs to be identified with precision. Alternatively, high turndown would be useful for process operations that switch from batch to continuous operation. In other cases, the same piece of equipment might be used on multiple applications of widely
10 varying energy release (or absorption) rates. Whilst the device previously described has considerable use it has its limitation for this type of application, because an impractical number of coils would be needed. Therefore an alternative embodiment of the invention uses a plurality of coils for varying the available heat transfer surface area as illustrated in example 3.

Example 3.

Table 4 sets out the heat transfer capacities of a series of coils of varying diameter and length.

Table 4

Coil diameter	Coil length range	Operating range
(mm)	(m)	(W)
1	0.13 – 0.6	4 – 22
2	0.9 – 2.3	40 – 141
3	1.9 – 4.7	118 – 429
4	3.0 – 7.8	249 – 913
5	4.4 – 11.8	457 – 1660

20

In order to arrive at the operating range, as with example 2, the LMTD is taken as 10°C and $(t_{\text{si}}-t_{\text{so}})$ as 10°C . The extremes of the ranges set out in columns two and three of table 4 represent the calculated values for minimum and maximum flow of the heat transfer fluid. Minimum flow is that which results in a pressure drop (of service fluid) of 0.1 bar.m^{-1} and maximum flow that which results in a pressure drop (of service fluid) 1 bar.m^{-1} .

This combination of coil diameters and lengths provides a system capable of very high turndown ratios. For example a six coil reactor can be designed to operate at less than 4 W and up to 5000 W. Table 5 shows the cumulative capacity of 6 coils of varying diameter.

Table 5

Coil number	Coil diameter	Coil range	Cumulative range
	(mm)	(W)	(W)
1	1 mm	4 – 22	4 – 22
2	1 mm	4 – 22	4 – 44
3	2 mm	40 – 141	4 – 185
4	3 mm	118 – 429	4 – 614
5	5 mm	457 – 1660	4 – 2274
6	5 mm	457 – 1660	4 – 3934

Each coil is sized for the maximum length shown in table 4. The nominal turndown ratio of the six coils is 984.

If $(t_{sr}-t_{so})$ is stepped down to 5°C when a single 1 mm diameter coil is operating, the nominal turndown ratio is increased to 1967 (2 – 3934 W).

If $(t_{sr}-t_{so})$ is stepped up to 20°C when all the coils are operating the nominal turndown ratio is increased to 3934 (2 – 7868 W).

The invention therefore enables a very large operating range with simple reactor design.

In some cases, rigorous analysis may require greater overlap (in terms of operating range) to ensure that pipes when opened can operate in the preferred fluid velocity range.

The invention can be used to improve the operation of commercial chemical and physical reaction systems. It can however also be used to provide considerably smaller

reaction systems with comparable commercial throughput. For example the invention enables reduction of reactor size by a factor of 10 and, in some instances, a factor of 100 or greater. In particular it can be applied to current commercial

- 5
 - batch organic synthesis reactions currently carried out in reactors of 10 to 20,000 litres.
 - bulk pharmaceutical synthesis reactions currently carried out in reactions of 10 to 20,000 litres.
- 10
 - batch polymerisation reactions currently carried out in reactors of 10 to 20,000 litres.
 - batch synthesis reactions of 10 to 20,000 litres currently used for unstable materials (compounds susceptible to self-accelerating runaways).
 - batch inorganic synthesis reactions currently carried out in reactions of 10 to 20,000 litres.

15

The techniques may also be useful in larger scale chemical and petrochemical operations.

CLAIMS

1. A reaction system in which a heat transfer fluid is carried through a process fluid in a conduit and the heat transfer surface area between the conduit and the process fluid can be varied according to the heat generated or absorbed by the reaction as determined by the temperature change in the heat transfer fluid across the reaction and the mass flow of the heat transfer fluid through the conduit.
2. A reaction system according to Claim 1 in which the conduit is made up of pipes or coils.
3. A reaction system according to Claim 1 or Claim 2 in which the conduit comprises of two or more heat transfer coils or pipes, which pass through the reaction fluid.
4. A reaction system according to Claim 2 or Claim 3 in which the wall of the pipes or coils are from $\frac{1}{2}$ to 4 mm thick.
5. A reaction system according to any one of the preceding Claims in which:
 - i. the average temperature difference between the heat transfer fluid and the processes fluid is from 1 to 100°C
 - ii. the temperature differential ($t_{sf}-t_{so}$) of the heat transfer fluid across the reaction system is at least 1°C
 - iii. the linear velocity of the heat transfer fluid is at least 0.1 metres/second.
6. A reaction system according to any of the preceding Claims in which the available surface area of the heat exchanger is sufficient to ensure that a temperature difference ($t_{sf}-t_{so}$) of more than 1°C is observed in the heat transfer fluid as it passes across the reaction.
7. A reaction system according to Claim 6 in which the temperature difference is more than 5°C.

8. A reaction system according to Claim 7 or Claim 8 in which the diameter to length relationship of a heat transfer coil is calculated by first calculating the heat transfer area required using the formula

$$U.A.LMTD = m.Cp.(t_{si}-t_{so}) \text{ (kW)}$$

5

where U = overall heat transfer coefficient (kW.m⁻².K⁻¹)

A = heat transfer area (m²)

m = mass flow rate of heat transfer fluid (kg/s)

LMTD = log mean thermal difference between service and process fluids (°C)

10

Cp = specific heat of heat transfer fluid (kJ.kg⁻¹.K⁻¹)

(t_{si}-t_{so})= temperature (°C) change in the heat transfer fluid between inlet and outlet

and the diameter to length relationship of the coil is developed to enable high Reynolds number in the heat transfer fluid without an excessive pressure drop.

15

9. A reaction system according to any of the preceding Claims comprising multiple heat transfer pipes or coils each of which has a diameter and length relationship designed to provide a certain degree of heat transfer and in which the pipes or coils may be brought into and out of operation according to the measured heat generated or absorbed by the reaction.

20

10. A reaction system according to any of Claims 2 to 9 in which the coils can be brought into and out of operation according to the heat transfer fluid flow requirements.

25

11. A reactor system according to any of the preceding Claims provided with control valves that regulate flow of heat transfer fluid to the heat transfer coils said valves being operated by an output signal from the measurement of the temperature of the reactor contents.

30

12. A reactor system according to Claim 11 in which the control valves are installed in series.

13. A reactor system according to Claim 11 in which the control valves are installed in parallel.
14. The use of a reaction system according to any of the preceding Claims for batch chemical synthesis reactions.
15. The use of a reaction system according to any of Claims 1 to 12 in batch organic synthesis reactions currently carried out in reactors of 10 to 20,000 litres.
- 10 16. The use of a reaction system according to any of Claims 1 to 12 in bulk pharmaceutical synthesis reactions currently carried out in reactions of 10 to 20,000 litres.
- 15 17. The use of a reaction system according to any of Claims 1 to 12 in batch polymerisation reactions currently carried out in reactors of 10 to 20,000 litres.
18. The use of a reaction system according to any of Claims 1 to 12 in batch synthesis reactions of 10 to 20,000 litres currently used for unstable materials (compounds susceptible to self-accelerating runaways).
- 20 19. The use of a reaction system according to any of Claims 1 to 12 in batch inorganic synthesis reactions currently carried out in reactions of 10 to 20,000 litres.
- 25 20. The use of a reaction system according to any of Claims 1 to 12 in a continuous reaction.



Application No: GB 0110299.5
Claims searched: 1-20

Examiner: Dave Mobbs
Date of search: 7 August 2001

Patents Act 1977
Search Report under Section 17

Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.S): B1X X16, X20; G3R RBQ59.

Int Cl (Ed.7): B01J 19/00; B01L 7/00; F28F 13/00, 27/00, 27/02.

Other: ONLINE: EPODOC, JAPIO, WPI, DIALOG DIALINDEX CHEMENG.

Documents considered to be relevant:

Category	Identity of document and relevant passage	Relevant to claims
X	US 5,762,879 (Todoroki Sangyo Kabushiki Kaisha) - see particularly figure 3 and column 6 line 67 - column 7 line 5.	1, 10, 12, 14, 21, 23, 25.
X	Collect. Czech. Chem. Commun., Vol 47 No 2, 1982, pages 446-453, Temperature control in a chemical reactor through variable area of the heat transfer surface. Experimental data. Horak J, Jiracek F, Jezova L.	1 at least.

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.

PUB-NO: GB002374948A
DOCUMENT-IDENTIFIER: GB 2374948 A
TITLE: Reaction system with variable heat transfer
surface area
PUBN-DATE: October 30, 2002

INVENTOR-INFORMATION:

NAME	COUNTRY
-------------	----------------

ASHE, ROBERT	GB
--------------	----

MORRIS, DAVID	GB
---------------	----

ASSIGNEE-INFORMATION:

NAME	COUNTRY
-------------	----------------

ASHE MORRIS LTD	GB
-----------------	----

APPL-NO: GB00110299

APPL-DATE: April 27, 2001

PRIORITY-DATA: GB00110299A (April 27, 2001)

INT-CL (IPC): B01J019/00 , G05D023/00

EUR-CL (EPC): B01J019/00 , F28F027/02 , G01N025/42